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INTRODUCTION

In March 2011 the final data from measurements for the ^{23}Na(n,n'γ) reaction were delivered to the CEA – Commissariat à l' Énergie Atomique, Cadarache, France in the context of the EURATOM-CEA collaboration agreement. This report documents that deliverable.

The measurement campaign was initiated in response to a request expressed by the CEA at a meeting of the Joint Evaluated Fission and Fusion nuclear data library project in 2007. This meeting took place under the auspices of the Nuclear Energy Agency (Organisation for Economic Co-operation and Development).

The CEA supports research for the advanced, Generation-IV type, sodium cooled fast reactor and is engaged in a project to develop a prototype: ASTRID – the advanced sodium test reactor for industrial development.

Inelastic scattering cross sections for sodium are of interest to the development of sodium cooled fast reactors. A recent OECD-NEA subgroup analysed the sensitivity of reactor parameters to cross sections and accordingly determined target uncertainties for the nuclear data [1]. Comparing these target uncertainties with the current status of nuclear data uncertainties and covariance data resulted in a list of target priorities. Among these features sodium inelastic scattering for which a target uncertainty of 4% was established for the average cross section in the energy range from threshold to 1.35 MeV. This is approximately seven times as good as the uncertainty for current evaluated data files for this isotope (see OECD-NEA High Priority Request List [2]).

At IRMM, the GAINS gamma-array for inelastic neutron scattering was developed with the purpose of measuring cross sections with uncertainties at or below the target uncertainties for nuclides like ^{23}Na using the (n, n'γ)-technique [3,4]. In response to the request, a measurement campaign of the ^{23}Na(n,n'γ) reaction was conducted with the GAINS array during 2009-2010, using metallic Na discs of 99.8% purity. The sample and the measurements were made at the Institute for Reference Materials and Measurements in Geel making use of GELINA, the Geel linear electron accelerator that drives a pulsed white neutron source allowing measurements by the neutron time-of-flight technique. A preliminary report of this work was presented earlier [5]. For the experimental work a careful review was made of the gamma-efficiency calibrations and the flux normalization in order to investigate in detail the corrections and the final uncertainties that may realistically be achieved.

DESCRIPTION OF THE EXPERIMENT

The present data were collected with the GAINS set-up (Gamma array for inelastic neutron scattering) [3] at the GELINA white neutron source for a total of 77 days during 2009-2010. The set-up is located at flight path 3 of the GELINA facility, at ≈ 200 m from the neutron source (198.7 m between neutron source and target).

GELINA was operated at 800 Hz repetition rate providing pulses of <1 ns FWHM. The incident energy of the neutrons was determined by the time of flight technique, using the time interval between the generation of neutrons at the source and the detection of the gamma rays produced by inelastic scattering in the target.

GAINS consists of 8 large volume HPGe Canberra detectors in angles of 110º and 150º to the beam, with 4 detectors positioned at each angle at distances of 14-16.5 cm from the target. The GAINS detectors have typical gamma energy resolutions of ≈ 2.3 keV for the 1.332 MeV peak of ^{60}Co. During the current measurement the performance of one of the 110º detectors was problematic resulting in significantly deteriorated energy resolution, and it was deemed necessary to reject all data from that detector.

The normalization of the measurements is provided by a ^{235}U fission chamber (FP3/200m) located upstream from the target (distance target – centre of FP3/200m: 147.5 cm), operated in parallel with the HPGe set-up. The fission chamber contains 3066(6) μg/cm^2 U in the form of highly enriched (99.827(4) %) $^{235}UF_4$ evaporated on 20 μm Al foils ([3], uncertainties in this report are one standard deviation total uncertainties). The fissile material is arranged in 8 layers (3 double-sided and 2 single-sided deposits) with a separation of 6 mm between electrodes (Figure 2). The chamber operates with P10 gas (a mixture of 10 vol.% methane and 90 vol.% argon) and provides a single output signal.

The data acquisition involved Acquiris DC440 digitizers with 12 bit resolution and a sampling rate of 440 MS per second. The system is described in detail in [4]. Data analyzed with the GAINS setup typically have a time resolution of ≈ 10 ns, corresponding to the sum (9.6 ns) of four time bins of the digitizers. This results in a neutron energy resolution of about 1 keV at 1 MeV.

Sample

The sodium metallic sample (TP-NP 08/10) [5] was prepared at IRMM from sodium ingot of elemental purity 99.8%, supplied by Alpha Aesar. The material was formed by cutting under argon atmosphere, rolling and punching under oil into a cylindrical disk of 79.80(8) mm diameter, 4.23(8) mm thickness and a mass of 19.44(4) g. Diameter, thickness and weight were determined following removal of the oil under argon atmosphere with low oxygen and water concentrations. In this atmosphere it was also enclosed in an Al can (0.50(1) mm wall thickness).
The sample had a density of 0.92 g·cm^{-3}, 95% of the theoretical density of metallic sodium (0.97 g·cm^{-3}). The critical parameter for the analysis is the mass per unit area 0.389(1) g·cm^{-2}, which follows from the measured mass and diameter. The sample diameter is larger than the beam diameter (61 mm).

Gamma-detector efficiency

The determination of the gamma detection efficiency was performed by a method combining calibration measurements and Monte Carlo modelling, as described in detail in ref. [6]. The calibration measurements were taken with a ^{152}Eu point source for a total of 38418(3) s. The total uncertainty associated with the ^{152}Eu source was 0.8%.
The measured efficiencies ($\varepsilon_{point,meas}$) of 11 ^{152}Eu gamma rays (245, 344, 411, 444, 779, 867, 964, 1112, 1213, 1299 and 1408 keV) were used to determine an MCNP5 model for each detector. This model was then applied to determine the extended sample efficiency for each observed ^{23}Na gamma energy E_γ, according to the expression:

$$\varepsilon_{ext}\left(E_\gamma\right) = \varepsilon_{ext,calc}\left(E_\gamma\right) \cdot \frac{\varepsilon_{point,meas}\left(E_\gamma^{Eu}\right)}{\varepsilon_{point,calc}\left(E_\gamma^{Eu}\right)} \qquad \text{(I)}$$

The ^{152}Eu gammas applied in (I) were E_γ^{Eu} = 444 keV and E_γ^{Eu} = 1408 keV, which were used to extrapolate to the ^{23}Na gamma energies E_γ of interest.
Taking into account the level of agreement between calculated (*calc*) and measured (*meas*) efficiencies, the overall uncertainties for the gamma detection efficiency were between 1.4-1.5% (Figure 1).

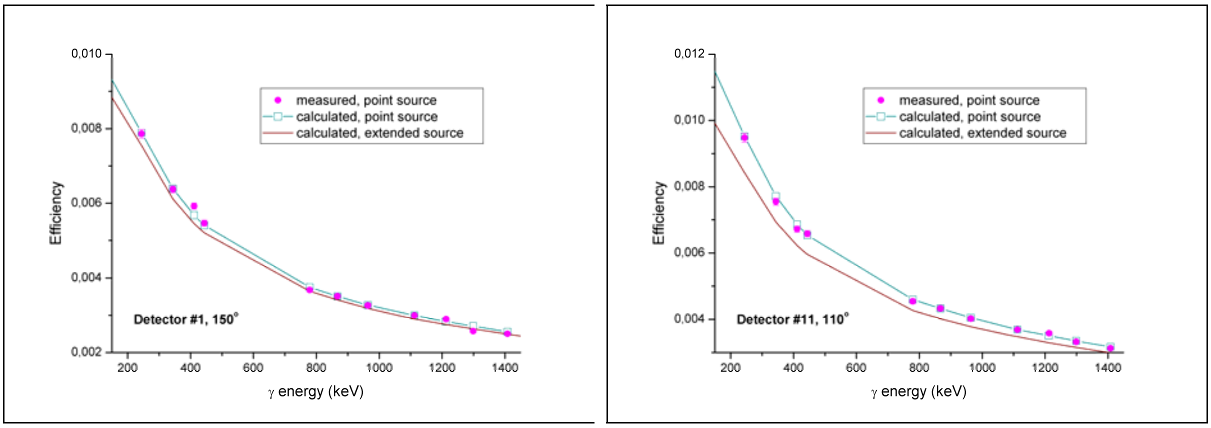

Figure 1. Measured and calculated (MCNP5) efficiencies for two of the GAINS detectors.

FP3/200m efficiency

The technique for the determination of the fission chamber efficiency that was used in [3] has recently been revised [5,7] and subsequently validated by an instrument intercomparison carried out at PTB [8]. The currently used method is based on the rejection of the alpha peak from the amplitude spectrum with the application of a threshold in the centre of the plateau between alpha and fission fragment peaks (Figure 2). A flat or linear fit of the plateau region is then extrapolated to zero pulse height (here considered to correspond with channel 0) to calculate the total number of fissions. Further corrections are applied to account for the polarity effect [7] of FP3/200m, the fission fragments that stop in the deposit [9] and the inhomogeneity of the UF_4 foils.
For the current measurement the selected threshold applied on the amplitude spectrum resulted in an efficiency of 83(1) %.

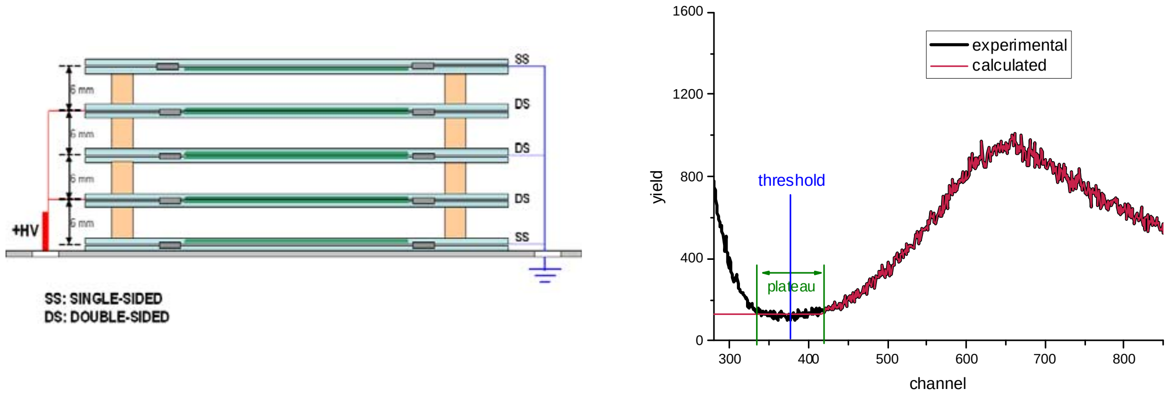

Figure 2. Left: schematic representation of the FP3/200m fission chamber. Right: experimental (black) pulse height distribution from FP3/200m and the calculated fission fragment spectrum (red).

Gamma production, level and total inelastic cross sections

The primary measured quantities with GAINS are differential γ-ray production cross sections, which result in total γ-production cross sections through angular integration [3]. These, in turn, give the level production cross sections with the use of level information from Firestone's evaluation [10], which is conveniently documented in ENSDF - the evaluated nuclear structure data file. Only one observed gamma per level is sufficient for this purpose. From these the total inelastic cross section can be produced for an energy region defined by the inelastic threshold and the highest observed transition.

In the current measurement, transitions up to the 6th level of ^{23}Na were observed, making the constructed total inelastic cross section reliable between the inelastic threshold (459.1 keV) to the threshold (3837.5 keV) of the 7th level with excitation energy 3677.60(4) keV.

Table 1 shows the observed gammas and associated levels of ^{23}Na(n, n'γ). The displayed information was obtained from the ENSDF Adopted Levels database [10]. According to Table 1, the contributions of the observed gammas to each level are related to the weights displayed in Table 2. The expressions involving these weights are found in reference [3].

E_γ (keV)	E_{level1} (keV)	$J_{\pi 1}$	$T_{1/2}$	E_{level2} (keV)	$J_{\pi 2}$	I_γ	γ mult.
439.986(10)	439.990(9)	5/2+	1.24(8) ps	0	3/2+	100	M1+E2
1635.96(3)	2076.011(22)	7/2+	24(2) fs	439.990(9)	5/2+	100.00(14)	M1+E2
1950.652(21)	2390.732(13)	1/2+	594(81) fs	439.990(9)	5/2+	52.1(8)	E2
2390.599(18)	2390.732(13)	1/2+	594(81) fs	0	3/2+	100.0(6)	
2639.70(5)	2639.85(4)	1/2-	58(11) fs	0	3/2+	100	
2263.39(3)	2703.500(25)	9/2+	95(4) fs	439.990(9)	5/2+	100.0(9)	E2 (+M3)
2541.92(4)	2982.060(19)	3/2+	2.5(4) fs	439.990(9)	5/2+	70.1(3)	M1+E2

Table 1: Observed gammas from ^{23}Na(n,n'γ) and associated initial (1) and final (2) levels [10].

γ-ray\Level	Total inelastic cross section	439.99	2076.01	2390.73	2639.85	2703.50	2982.06
439.99	1.000	1.000					
1635.96	0.089(1)	-1.000(2)	1.089(1)				
2390.60	1.000	-0.521(6)		1.521(9)			
2639.70	1.000				1.000		
2263.39		-1.000(9)	-0.544(8)			1.544(10)	
2541.92	1.427(7)	-1.000(5)		-0.007(2)			2.434(8)

Table 2: Contributing weights of the observed gamma production cross sections to the construction of the level cross sections (See reference [3] for a description of the use of these weights).

First results for the two lowest levels, obtained with part (≈ 17 days) of the current dataset, have been presented in [5]. Those were obtained with preliminary efficiency calculations and without the application of multiple scattering corrections.

The current results are produced from the full dataset (966.2 hours) and are corrected for multiple scattering and attenuation effects. These corrections were calculated with the help of iterative MCNP5 simulations, using the γ-production cross sections for ^{23}Na obtained from the current measurement. Convergence was obtained in a few steps.

PRESENTATION OF RESULTS

The total inelastic scattering cross section for ^{23}Na is shown in Fig. 3, displayed for the energy region between the inelastic threshold (459 keV) and the threshold of the 7th excited state (3837.5 keV).

A comparison with some of the existing data shows excellent agreement with the data of Kopecky et al. [12] up to 1600 keV, and overall good agreement with earlier data from Perey et al. [11].
With respect to the evaluated nuclear data libraries, the structure displayed in the lower energies is very well described by JEFF-3.1 up to about 1600 keV. Good agreement is also observed with ENDF/B-VII up to ≈ 2200 keV. Beyond that energy there are very few experimental points available and the current measurements show comparable deviations from the evaluated inelastic scattering cross sections of the two databases.

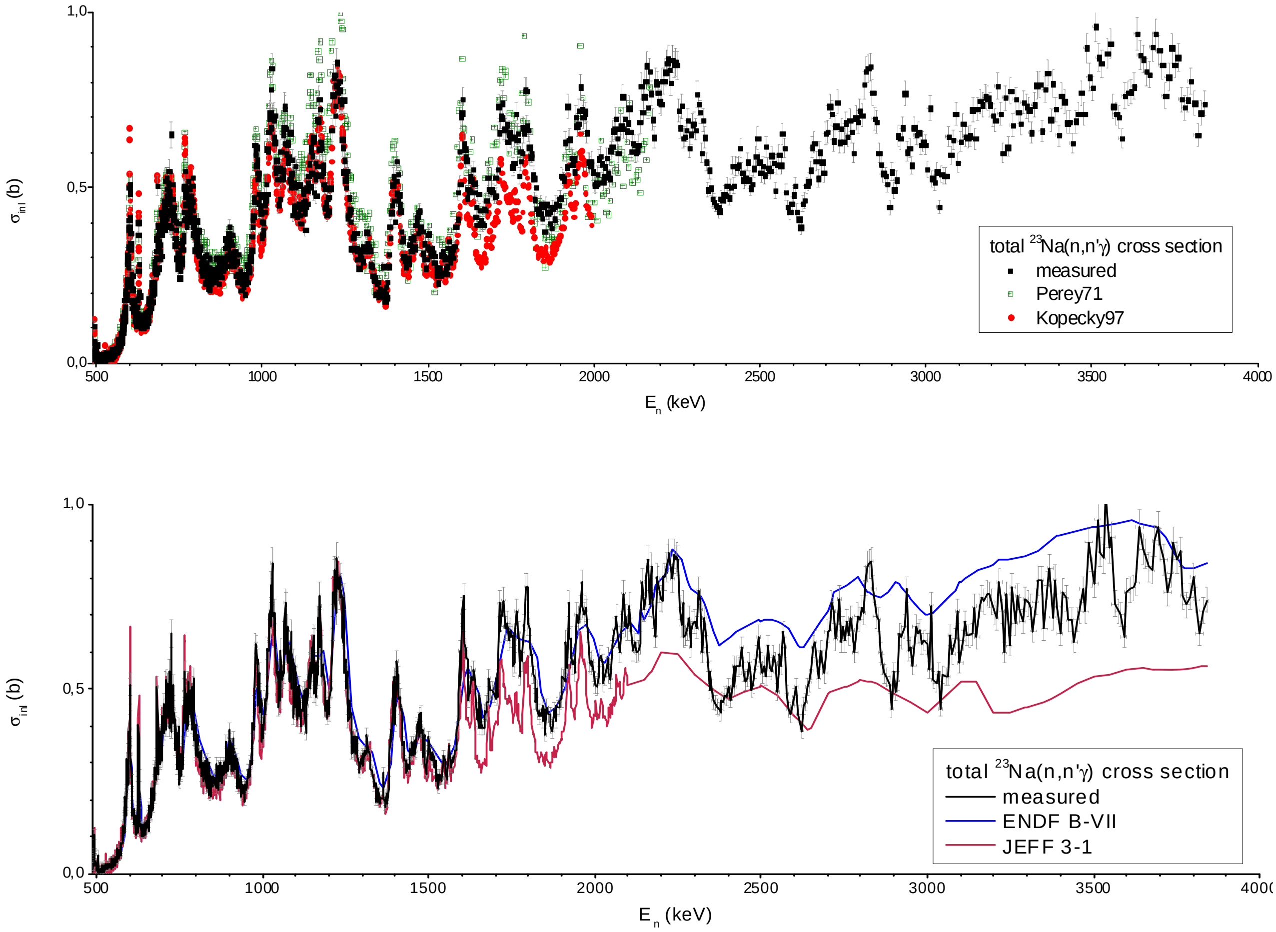

Figure 3. The measured total inelastic cross section for ^{23}Na (0Lxs.his file), compared with earlier measurements ([11, 12]) (top) and evaluated data from the ENDF B-VII and JEFF 3.1 libraries (bottom).

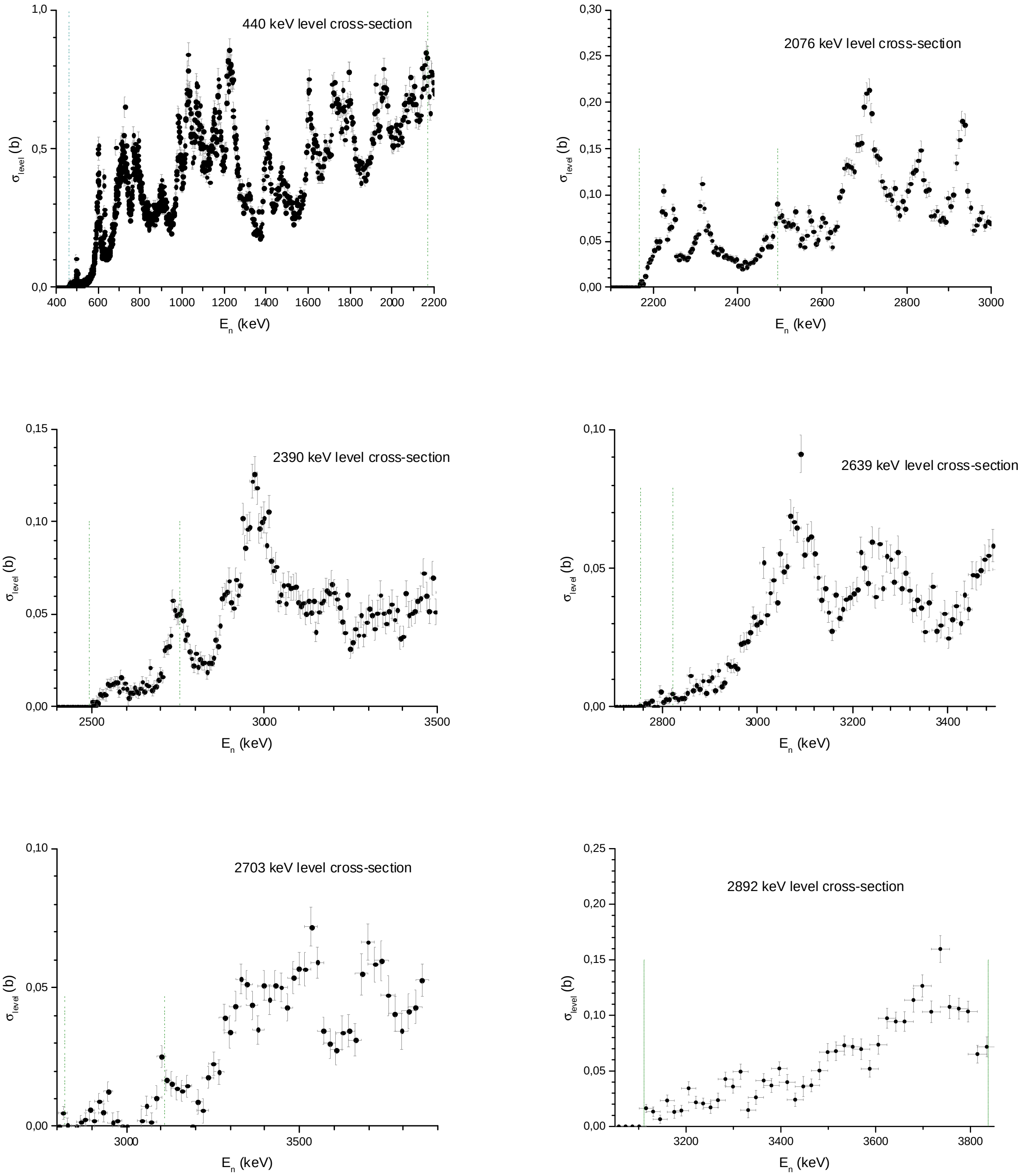

Figure 4. The level production cross sections for the first six levels of ^{23}Na, given in the nnnnLxs.his files. The dotted lines indicate the thresholds of the displayed and of the next higher level.

THE DATA AND HOW TO USE THEM

The delivered data files contain the total inelastic cross section and the level cross sections of the first six excited states of ^{23}Na. Due to the principle applied in the measurements (the (n,n'γ)-technique), the maximum incident neutron energy for which these are valid is E_n = 3837.5 keV, which is the threshold energy for excitation of the seventh excited state. Beyond this energy another source of information should be used (existing JEFF-3.1.1 for instance).

File 0Lxs.his is the total inelastic cross section (Figure 3), which corresponds to MT number 4 of the ENDF format. Graphs of this cross section and a comparison with earlier work can be found in the file total_inelastic_xs.xls.

The nnnnLxs.his files (Figure 4) with nnnn>0 are cross sections for population of the levels with excitation energy nnnn keV. These files correspond to MT numbers 51, 52, 53, 54, 55 and 56 of the ENDF format.

Each .his file has four columns. The first is the time-of-flight in ns, the second the energy in keV, the third the cross section in b and the fourth the uncertainty of the cross section in b.

Summary

The final inelastic scattering data obtained at IRMM with the GAINS setup at the GELINA time-of-flight facility were delivered to CEA in the form of data files valid up to an incident neutron energy of 3837.5 keV. Special attention was paid to a re-evaluation of the analysis procedure and the evaluation of experimental uncertainties. Two uncertainties affect the data at all energies uniformly; the gamma detection efficiency of about 1.5% uncertainty and the efficiency of the fission chamber with an uncertainty of about 1.3%. The remaining contributions to the uncertainty are from the ^{235}U standard fission cross section [13] and from statistics. The latter source of uncertainty is fully uncorrelated. In view of the large number of measured points in the energy interval from threshold to 1.35 MeV the present data meet the target uncertainty of 4% on the average cross section in this interval [2]. The estimated uncertainty for the average total inelastic cross section of the present data for this energy interval is 2.3%

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Abstract
This report describes the inelastic scattering data delivered to CEA, March 2011. The GAINS setup was used for measurements of the inelastic neutron scattering of Sodium using the (n, n'γ) technique. The experimental work was performed at the GELINA facility at a 200 m flight path with eight high purity germanium detectors, using a metallic sodium sample prepared at IRMM.
A brief description of the experimental details and the final results for the total inelastic and level production cross sections are presented.

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